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widths of a few MHz. Whether or not these processes can reach threshold depends upon the resolution of basic issues lying in an interdisciplinary region between quantum electronics and nuclear physics that have not been previously addressed. It was the purpose of this work to study these issues experimentally.

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ANNUAL SUMMARY REPORT

for the period

1 January 1987 through 31 December 1987

for

Office of Naval Research
Contract N00014-81-K-0653
Task No. NR 395-072

THE DEMONSTRATION OF THE FEASIBILITY OF THE
TUNING AND STIMULATION OF NUCLEAR RADIATION

Short Title: GAMMA-RAY LASER

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PROJECT DESCRIPTION

This project concerns the demonstration of the feasibility of the tuning and stimulation of nuclear radiation. It represents a critical line of investigation in our overall program concerned with the feasibility of a gamma-ray laser.

Theory, supported by our experiments conducted under this contract, has indicated that anti-Stokes Raman upconversion of intense but conventional laser radiation produced by scattering from isomeric states of nuclear excitation could lead to significant sources of tunable gamma radiation characterized by the natural Mössbauer widths of the lines. Further computations have suggested that this type of coherent, as well as a type of incoherent, optical pumping could even lead to appreciable levels of inversion of the populations of nuclear levels, thus supporting the growth of stimulated gamma-ray intensities. Whether or not these processes can reach threshold depends upon the resolution of basic issues in an interdisciplinary region between quantum electronics and nuclear physics that have not been addressed elsewhere. It is the purpose of this contract work to study these issues experimentally in order to guide the development of the technology and methods needed to exploit the enormous potential of this effect.

SCIENTIFIC PROBLEM

The viability of the concept for the tuning of gamma radiation by adding the variable energy of an optical photon produced by a tunable laser depends upon the existence in the nucleus of a particular arrangement of excited states. A suitable energy difference would make it possible to dress the nuclear states with the laser photons. Transitions between the dressed states would then occur at the sum and difference

frequencies characteristic of the nuclear transition, plus or minus the energies of integral numbers of laser photons.

Whether the necessary arrangements of nuclear states do exist is the central issue being addressed in this contracted work. Surprisingly, such information is currently unknown because such potentially useful states would lie in the "blind spots" of conventional techniques of nuclear spectroscopy. Normal Mössbauer spectroscopy provides enormous resolution, but a tuning range that is inadequate by orders of magnitude to support any possible study of transitions to the intermediate states of a multiphoton process. Conversely, crystal spectrometers provide broad tuning ranges, but levels of resolution that miss by two orders of magnitude the threshold that would be necessary to separate the transitions to the initial and intermediate states. As a consequence, the ideal arrangement of nuclear energy levels needed for the Raman upconversion process could be a common occurrence that has gone unnoticed because of the inadequacies of conventional nuclear spectroscopy.

The critical problem in this research has two facets: 1) the development of an appropriate spectroscopic technique, and 2) the search for a suitable medium for a large-scale effect. The dressing of the nuclear states not only affects their energies, but also changes their transition properties. Forbidden nuclear transitions should become allowed so that the metastability of isomeric states would be "switched off" as the states were dressed. This would greatly enhance the prospects for stimulating the gamma-ray transition, in addition to rendering it tunable. It is the development of the investigative instrumentation and the verification of these predicted effects that comprise the scientific problem addressed by this contract research.

TECHNICAL APPROACH

For the resolution of the central issue of the existence of potentially useful intermediate states in a multiphoton upconversion of optical photons to gamma-ray energies, it was first intended to demonstrate sum frequency generation in one case in which nonresonant intermediate states were known to exist. This was the case in which both initial and intermediate states were magnetic sublevels of the same nucleonic state and in which the transitions were mediated by the $M1$, magnetic dipole operator. Experimental data reproduced in the literature suggested that such a process had already been unknowingly demonstrated for the generation of radiofrequency sidebands to Mössbauer transitions at the sum and difference frequencies. This suggested the development of a new instrument, a Frequency Modulation Spectrometer for gamma-ray energies, designed to support the needed studies of nuclear structure with the precision of Mössbauer spectroscopy applied over a tuning range of energies lying considerably beyond the state-of-the-art at the time our work began. With this instrument, we are conducting Mössbauer experiments in the presence of intense radiofrequency fields with measurement and parameterization of the conversion efficiency into the sum frequency lines to determine the practical limits on the ultimate linewidths and tuning ranges that can be achieved. This technique will then be used in a "bootstrap" approach to support a search for accidentally resonant intermediate states. By replacing the radiofrequency excitation with tunable higher frequencies, it is expected that the tuning range of Mössbauer spectroscopy can be extended by further orders-of-magnitude.

PROGRESS DURING THIS REPORTING PERIOD

Instrumentation of FMS

The sum and difference frequency sidebands produced on intrinsic Mössbauer transitions have made possible very effective new instrumentation for high resolution spectroscopy at gamma-ray energies. A prototype version of this Frequency Modulation Spectrometer (FMS) was first described¹ by our laboratory in 1985, and subsequent refinements were made during the successive reporting periods under this contract. This device monitors changes in the intensity of transmitted single-frequency gamma photons as a function of frequency of the long wavelength photons of the alternating magnetic field in which the absorbing nuclei are immersed.

With this contract support our prototype "Nuclear Raman Spectrometer" was refined into mature technology resulting in the Frequency Modulation Spectrometer, (FMS) for gamma-ray energies². The original prototype device had required a tedious level of manual interaction, and this was replaced with a fully-automated and computerized control system. At its heart is a multi-channel scalar (MCS) and IEEE-488 GPIB interface with an Apple II+ computer. The MCS was designed to have a 100% duty cycle. The GPIB enables the spectrometer to sweep continuously through the frequencies of an rf magnetic field with a Wavetek frequency synthesizer. The Mössbauer drive allows the frequency of the gamma photons to be biased by a constant Doppler shift, if desired. In its present form, the FMS device has an instrumental resolution of 100Hz and a continuous tuning range of 10⁹Hz with a stability of 0.1Hz/sec with no mechanical movements required anywhere. These characteristics are comparable to a Mössbauer spectrometer with a means of shifting the gamma-ray source, having a resolution of 10nm/sec and a range of 100mm/sec with a stability of 0.01nm/sec/sec. Demonstration spectra were acquired with ⁵⁷Fe showing isomer shifts and thermal shifts. Because we were using a modulation

type of spectroscopy, the static features could be suppressed, and these different effects were obtained with unprecedented clarity.

Details of the mature FMS device are contained in the article "Frequency-modulation spectrometer for Mössbauer studies", by P. W. Reittinger, T. W. Sinor, S. S. Wagal and C. B. Collins, Rev. Sci. Instrum. 59, 362 (1988). A copy is reproduced in Appendix I and shows data demonstrating the remarkable detail which can be readily obtained with this device. The crowning achievement is found in Fig. 6 of that reprint where 24 sidebands are seen in a tuning interval of 50 MHz, all having values of relative intensity confirmed in the companion plot of a computer synthesis of that region of the spectrum.

It appears that the FMS device developed under this contract has now proven its utility as a tool in Mössbauer spectroscopy. Besides standing ready to serve in our own search for nearly resonant intermediate states for a multiphoton process, it has the versatility to support many other kinds of experiments as well.

Spurious Acoustic Effects

Despite the impressive agreement between results obtained with the new instrumentation developed under this contract and rudimentary multiphoton models^{2,3} intense criticism has continued to issue because of the belief that, somehow, all effects which appear as sidebands arise from spurious acoustic vibrations excited by magnetostriction of some of the various elements of the spectroscopic sample and its holder. In appearance our multiphoton spectra resemble the transmission spectra which Ruby and Bolef⁴ obtained by imposing periodic Doppler shifts of purely mechanical origin upon the Mössbauer source. This unfortunate similarity in appearance between phenomena arising from such different origins has provided the basis for years of critical controversy.

There is the disturbing impression in the Mössbauer community that an acoustic origin had been "proven" for all sidebands by the benchmark

experiment of Chien and Walker⁵ in 1976. In that experiment an absorbing foil composed of ferromagnetic and nonmagnetic layers was used to study transport of the causative agent from the ferromagnetic layer into the nonmagnetic region where the sidebands were produced upon Mössbauer transitions of embedded ⁵⁷Fe nuclei. Very clear evidence showed that the cause did arise in the ferromagnetic Ni layers, producing sidebands in the nonmagnetic stainless steel layers. The most ready explanation at that time was a transport of phonons from one layer to the next with a high acoustic Q. Those experiments were repeated in the current reporting period⁶ but with extensions which contradict the classic interpretation of Chien and Walker.¹ In fact, our reexamination shows the original experiment to have been so flawed that any conclusions drawn from it now must be considered unproven.

As detailed in the manuscript⁶ "Comment on Mössbauer sidebands from a single parent line," by C. B. Collins, P. W. Reittinger, and T. W. Sinor, submitted to Phys. Rev. B, sideband effects scale with the square of the number of ferromagnetic sources. Interpreted as indicating the addition of fields from each source that are squared to communicate power into the sideband intensities, such scaling is completely inconsistent with an acoustic origin. In experiments such as these, acoustic phonons are the bosons associated with vector fields driven by tensor forces, not vector forces. Without invoking stimulated emission, we can conceive of no way in which tensor sources which are physically separated can produce coherent vector fields in a space between them, even if they are temporally synchronized. The stimulated emission of phonons to produce coherent additions of the displacements arising from the different sources would imply the existence of a threshold of power, above which two modulation indices of m would give an effect of $4m^2$ and below which only $2m^2$. No such threshold was suggested by the data which was obtained

over an adequate range of powers. Sidebands scaled consistently with $4\pi^2$.

At the time of reporting a critical experiment was initiated which will go much further in eliminating the possibilities of an acoustic origin to the large scale Mössbauer sidebands we observe by studying the transport of the causative effect over greater distances than the thickness of a layer. However, at this juncture it can already be said that the experiment of Chein and Walker which is considered to be the bulwark of the acoustic model for Mössbauer sidebands actually showed nothing but a fortuitious arrangement of errors.

SIGNIFICANCE

The significance of the work completed during this past year is twofold. The first arises from completion of the FMS instrument which now stands as a powerful tool for use in the search for accidentally resonant intermediate states needed to dress nuclear states with photons to an extent necessary for large scale effects. The second significance accrues from the increased confidence in the identification of Mössbauer sidebands in magnetic media as manifestation of the successful excitation of dressed states. Destruction of the "proof" that all such large effects were caused by spurious phonons leaves the field open for other explanations. Indeed, the dressed state model is adequate to explain the phenomena but had never been proven necessary. When another explanation was available it had been the extraneous alternative.

Now there is no longer any proof or even evidence of the general pervasiveness of acoustic sidebands in magnetic materials. The next experiments emphasizing transport of sidebands should make a definitive statement about the origins of radiofrequency sidebands in Mössbauer spectroscopy.

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Appendix I

"Frequency modulation spectrometer for Mössbauer studies," by P. W. Reitinger, T. W. Sinor, S. S. Wagai, and C. B. Collins, Rev. Sci. Instrum. 59, 362 (1988).

Frequency-modulation spectrometer for Mössbauer studies

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A nuclear frequency-modulation spectrometer (NFMS) for high-resolution gamma-ray spectroscopy is described in this article. As the name implies, this device operates by modulating the cross section for gamma-ray absorption. The automation of this spectrometer required the development of an interface to an Apple computer which provides a real-time data display. This interface also enables the Apple computer to control up to two Mössbauer spectrometers at once, with a real-time data display for each. A nuclear frequency-modulation spectrometer makes it possible to observe directly the phenomenon known as "rf sidebands" in Mössbauer spectroscopy, without interference from the "parent transitions." The high resolution of NFMS makes it possible to examine the "rf sidebands" for any fine structure.

INTRODUCTION

As early as 1960 it had been noted that radio-frequency (rf) sidebands to the hyperfine structure of ^{57}Fe could be observed with a Mössbauer spectrometer.¹ The six lines (parent transitions) in a normal absorption spectrum of ^{57}Fe in iron [Fig. 1(a)] are accompanied by additional absorption peaks (rf sidebands) when the absorber is subjected to a rf field [Fig. 1(b)]. In 1960, Ruby and Bolef reported the observation of rf sidebands in iron produced by mounting a ^{57}Co Mössbauer gamma-ray source on an ultrasonic transducer driven at MHz frequencies.¹ It should be noted that the rf transducer was used in addition to a long-period oscillator which provided the energy range for the Mössbauer spectrum by introducing controlled Doppler shifts. In 1968, Perlow reported the generation of rf sidebands in iron directly, by subjecting the gamma ray source to a rf field without the involvement of any external ultrasonic source.² In that same year, Heiman, Pfeiffer, and Walker reported observing rf sidebands in iron as a result of subjecting the iron foil absorber to a rf field.³ Finally, in 1976, Chien and Walker presented a method for producing rf sidebands in a nonferromagnetic stainless-steel absorber with a rf field, by using nickel as a ferromagnetic nonabsorbing driver.⁴ In all cases, the rf sidebands appeared at integral multiples of the frequency of the applied rf (Fig. 2). Figure 2 shows rf sidebands produced in a stainless-steel foil driven by a nickel foil immersed in rf fields of different frequencies. The frequency dependence of these rf sidebands can be utilized to make a high-resolution adaptation of Mössbauer spectroscopy which is freed from many of the mechanical constraints tending to limit conventional devices.

In 1967, Bolef and Mishory reported the development of a spectrometer which was based upon rf sidebands induced in a Mössbauer source with a rf electromechanical transducer (an X-cut quartz crystal).⁵ As the frequency of the applied rf was changed, the energies of the sideband gamma-ray emissions changed. This phenomenon enabled Bolef and Mishory to obtain an absorption spectrum as a function of the frequency of the applied rf. In 1985, DePaolo, Wagal, and Collins reported success in developing a spectroscopic

technique using rf sidebands induced in a ferromagnetic absorber by a rf field.⁶ Modulating the absorber has numerous advantages over modulating the source. It is easier and safer to work with a stable isotope, and it is also easier to interpret a spectrum from a single line source, as opposed to a Zeeman split source or a source with rf sidebands. Therefore, we have improved the technique for a modulated gamma-ray absorption cross-section spectroscopy which we call nuclear frequency-modulation spectroscopy (NFMS).

The technique reported by DePaolo, Wagal, and Collins was slow and laborious, with data collection times on the order of months for tens of data points. We would like to report the automation of this technique, with resulting data collection times of two days for 1024 data points and a sig-

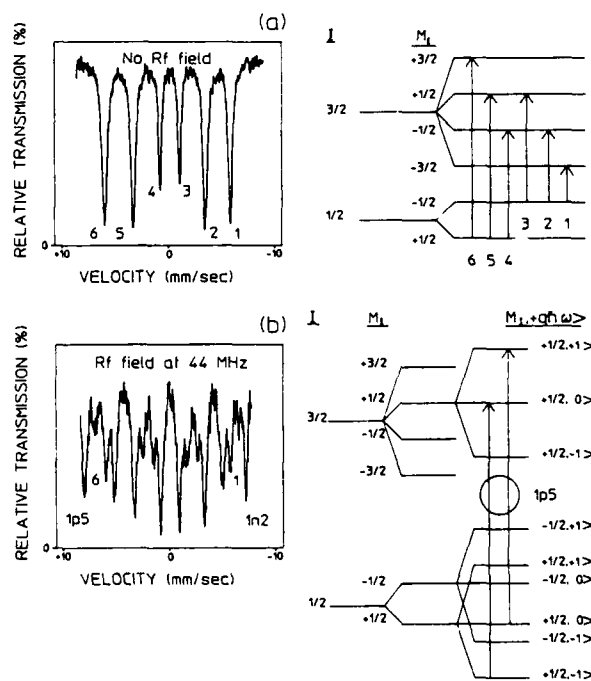


FIG. 1. Mössbauer absorption spectra and energy level diagrams for ^{57}Fe in iron; (a) with no rf field at the absorber, and (b) with a 4-Oe rf field applied to the absorber at a frequency of 44 MHz, showing the effect of the rf field.

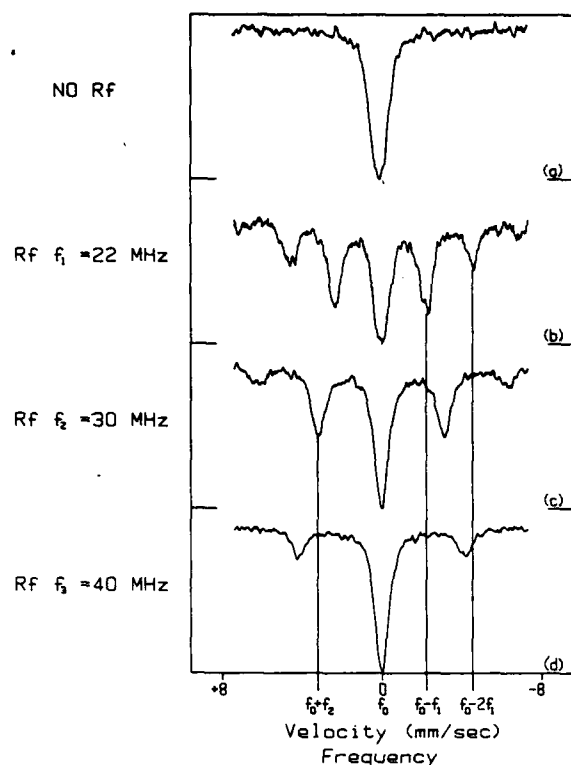


FIG. 2. Mössbauer absorption spectra of ^{57}Fe in 310-stainless-steel driven by a nickel foil, showing the effect of a rf field applied to the absorber-nonabsorber sandwich at frequencies of (b) 22 MHz, (c) 30 MHz, and (d) 40 MHz in comparison to (a) a no rf spectra. It can be seen from the figure that sidebands appear at integral multiples of the frequency of the rf field.

nal-to-noise ratio of 8:1 for a signal that represents a relative absorption of 3%. This article describes the NFMS apparatus, describes the interface to an Apple computer which automates NFMS data collection, and presents some typical NFMS data.

I. SPECTROMETER DESIGN

The NFMS is a modification of a conventional Mössbauer spectrometer comprised of the equipment in the dotted box in the schematic of Fig. 3. A Kr gas-filled proportional counter (ASA PC-KR-1) biased with 1.8 kV from a Bertran Associates model 303 dc voltage supply was used as our gamma-ray detector. The signal from the detector was amplified by an ASA CSP-400A preamp and ASA LA-200 amplifier. The amplified signal was then fed into an ASA LG-200 linear gate which produced 1- μs TTL pulses for counting.

A 10-mCi ^{57}Co Mössbauer source in a Pd matrix was mounted on an ASA K-4 linear motor capable of operating at a constant velocity or with constant acceleration. A stable means of Doppler shifting the energy of the emitted gamma ray is needed, therefore, an ASA S-700 motor controller is used to produce the voltage waveforms which drive the linear motor. The constant acceleration voltage waveform is derived from a 5-Hz square wave which must be provided by the multichannel scalar (MCS). If the motor is driven at a constant velocity, then the motor controller gates off data to the MCS while the motor is rewinding.

The key to NFMS is the presence of the rf field at the absorber, for which a very stable rf signal generator and amplifier are needed. A Wavetek 3510 signal generator, with a frequency range of 1 MHz to 1 GHz, and a 100-Hz resolution with a 500-Hz/(10-min) stability, was used. The rf amplifier was an ENI 550L 50-W linear amplifier with a range of 1.5–400 MHz. There were two basic circuits used to generate the rf field at the absorber. One was a series LC circuit in parallel with an impedance matching capacitor (Fig. 3). This series-resonant tank circuit was designed to have either a low Q when used in a narrow-band NFMS, or a high Q when used to obtain a Mössbauer spectrum in the presence of a single-frequency rf field. It should be noted that narrow band in these instances refers to a 12 MHz or less bandwidth.

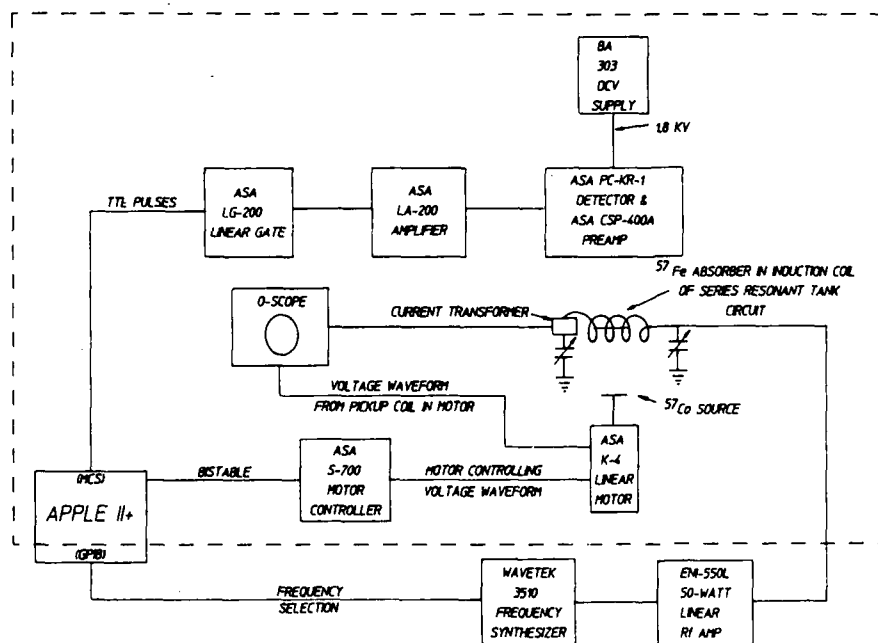


FIG. 3. This schematic shows the NFMS, while the portion of the apparatus which is in the dotted box can be used as a conventional Mössbauer spectrometer.

The other circuit, used in the wideband NFMS, simply incorporated an inductor in series with an impedance matching noninductive load. The absorber was then mounted in the induction coil of the appropriate circuit and subjected to field intensities on the order of 1–5 Oe.

In order to monitor the field intensity in the coil, a Pearson 2877 current transformer was used to measure the current flowing into the induction coil. This transformer outputs 1 V/A with a usable range of 300 Hz to 200 MHz and an insertion impedance of 0.02 Ω . In order to monitor the velocity of the motor, there is a pickup coil mounted in the linear motor. The output from this pickup is used to stabilize the driving voltage waveform, but it can also be monitored on an oscilloscope. The velocity of the motor was established by correlating the pickup coil voltages to the positions of the peaks in the six-line spectrum of ^{57}Fe . Currently, work is underway building an interface to an Apple II+ from an ASA LC-9A laser interferometer. This interface will enable the computer to display real-time velocity information as well as track any drifting.

The heart of the NFMS, however, is the MCS/GPIB interface (Fig. 4). It enables an Apple II+ computer to be used for data acquisition and real-time data display with either the conventional Mössbauer spectrometer (constant acceleration mode) or with the NFMS (constant velocity mode with GPIB interface to signal generator). The MCS is a card designed around two VIAs, or versatile interface adapters (6522's). The GPIB, or IEEE-488 General Purpose Interface Bus (9914), is a commercially available interface card available for the Apple computer. The GPIB is necessary only for scanning frequencies of the signal generator. Therefore, the GPIB is not needed if one intends to use only the Mössbauer spectrometer.

The central components of the MCS are the two 6522's, the multiplexing logic, and a 12-bit counter. Each 6522 is a

40-pin chip which has a 16-bit counter with a 16-bit latch, a 16-bit counter with an 8-bit latch, two 8-bit parallel ports, and a serial port. The counter with the full latch can be set to count down in a free running mode and generate interrupts. In other words, the 6522 can be set to generate evenly spaced interrupts so that the Apple's CPU need not be wasted keeping track of time. The counter with the half-latch can be set to count negative logic pulses at one of the pins of the 6522. The multiplexing logic is an assortment of gates which channel the pulses to be counted to one of the 6522's while channeling the Apple's data bus to the other 6522. When an interrupt is generated, the pulses to be counted are gated to the other 6522 while the Apple's data bus is then channelled to the first 6522. As a result, the time it takes the Apple's CPU to add a count to the proper channel is not dead time for the MCS. The 12-bit counter, actually three 4-bit counters, is needed to count 512 interrupts. This counting produces the 5-Hz square wave which is used by the motor controller to generate a constant acceleration voltage waveform for the linear motor. Therefore, the time between interrupts, hence the dwell time per channel, must be 195 μs for a 1024 data point Mössbauer spectrum. The NFMS, on the other hand, does not require an accelerating source. Therefore, when using the MCS in a NFMS, the dwell time can be user selected. The optimum dwell time minimizes the total dead time, which arises from the time needed to allow the rf signal to stabilize each time the frequency is changed, without compromising the stability of the signal.

Use of this hardware as a NFMS or a Mössbauer spectrometer is determined by the software. Written in 6502 assembly language, the software for the two spectrometers is similar in principle but different in particulars. In both spectrometers the MCS transfers data to the Apple on an interrupt basis. Both programs consist of four basic routines: an initialization routine, a display routine, a keyboard-inter-

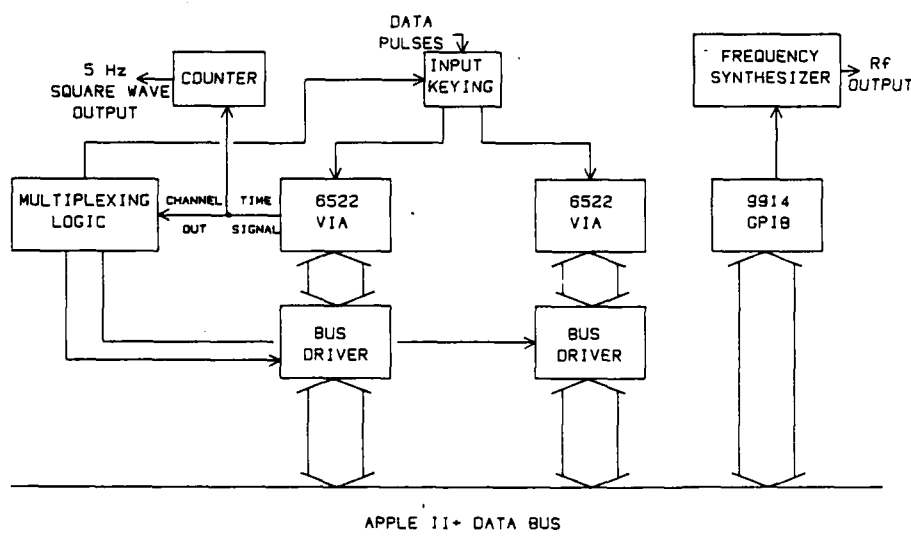


FIG. 4. This block diagram shows the basic components of the interface to the Apple II+ computer. This interface enables the computer to be used for automatic NFMS data acquisition, or it can enable one computer, with two cards, to control and collect data from up to two Mössbauer spectrometers.

MCS/GPIB INTERFACE

preparing routine, and an interrupt routine. The initialization routine uses the multiplexing logic on the MCS card to address each of the 6522's and set the appropriate registers. The display routine has two options. The data can be displayed graphically at different resolutions, or counts can be displayed as counts per channel and total counts per sweep. The graphics data display uses table lookup and two graphics screens to provide a real-time data display. The initialization routine generates a table in memory which stores the address for a given vertical coordinate on the screen in a memory location which is correlated to the value of the vertical coordinate. The value of the horizontal coordinate is correlated to the memory address of the channel to be displayed. As the display routine scans through memory at the data, the datum value and the channel value are used to address indirectly the appropriate graphics screen coordinate through this table. Therefore, by using table lookup, all of the mathematical operations necessary to obtain the appropriate screen addresses (including a division by seven) are performed only once. While one graphics screen is displayed, the other is cleared and plotted with the current data. The updated screen is then activated and the first screen is cleared and replotted, and so on. The keyboard-interpreting routine allows one to change the display, change the resolution of the graphics display, or stop the spectrometer and store the data on a disk, all with single key codes. A table of these key codes is displayed at the bottom of the counts display screen, the default display. Finally, the interrupt routine collects the count from the currently accessible 6522 and stores it in the appropriate 3-byte location. The display and keyboard routines for the two spectrometers are identical, but the initialization and interrupt routines for the two spectrometers are necessarily very different.

In the Mössbauer spectrometer, the initialization routine must enable the MCS card to generate interrupts at 195- μ s intervals. Next, the interrupt routine must be capable of pushing all values in the CPU's registers to the stack, accessing the count from the appropriate 6522 and adding it to the appropriate memory locations, and then reloading the CPU's registers with their initial values, all in less than 195 μ s, with enough time left over to update the display between interrupts. This feat was best accomplished by using four separate interrupt routines. Since the spectrometer has 1024 channels, the data is stored in twelve 256-byte pages for 3 bytes per channel. Each interrupt routine addresses a channel comprising 3 bytes through the sum of base addresses plus a counter value. Upon completion, each routine stores the address of the next interrupt routine in the interrupt vector. The fourth routine stores the address for the first routine in the interrupt vector and increments the addressing counter. The result is an interrupt routine that lasts 50–60 μ s from interrupt to return, depending on the number of bytes which must be incremented. As a result, one Apple II + computer can easily handle two Mössbauer spectrometers with a real-time data display for each.

In the NFMS, the initialization routine enables the MCS card to generate interrupts at $\frac{1}{60}$ of a second intervals. This routine also initializes the signal generator through the GPIB. The interrupt routine must then translate the number

of interrupts generated into an elapsed time and compare this time to the selected dwell time. In addition, this interrupt routine must perform all of the functions of the Mössbauer spectrometer interrupt routine. After the elapsed dwell time, the interrupt routine must step the frequency of the signal generator and change the address (channel) for data storage. When the frequency of the signal generator is changed, and for a time thereafter, the data to the MCS must be gated off and the timing stopped until the signal is stabilized. In the NFMS, the speed of the interrupt routine is no longer a major concern due to the significant increase in the time between interrupts and the fact that the data is gated off while the interrupt routine is delaying for the signal generator. Unfortunately, however, the time required to change the frequency is unavoidable dead time. Yet the total dead time in a run can be minimized by selecting a sufficiently long dwell time which does not allow the signal to drift significantly.

II. DATA AND DISCUSSION

Figures 1 and 2 show data collected with our Mössbauer spectrometer and processed with a five-point running average. These figures show rf sidebands in an iron foil absorber, and the frequency dependence of the rf sideband energies in a stainless-steel foil absorber. All NFMS spectra to be shown were obtained from a 1.5-cm \times 0.85-cm \times 2.5- μ m iron foil absorber enriched with 95% ^{57}Fe . This foil is the same absorber which gave us the spectra in Fig. 1. All spectra shown were obtained from an absorption geometry, using a ^{57}Co source in a Pd matrix. All NFMS spectra have been processed with a five-point running average.

The first set of NFMS data concentrates on the first-order sidebands from the 1 and 6 parent transitions (Fig. 5). The nomenclature for identifying the rf sidebands is as follows. The first digit corresponds to the order of the sideband. Radio-frequency sidebands of the j th order from a given parent transition are found at the sum and difference frequencies of the static field, or Zeeman splitting, and j times the frequency of the applied rf field. The letter after the first digit, either an "n" or a "p," indicates whether the sideband is a negative or positive sideband, respectively. A negative sideband appears at an energy lower than the energy of the parent transition, while a positive sideband is at a higher energy. The last digit identifies the parent transition of the sideband. There are six allowed transitions for ^{57}Fe in a metallic iron foil, of which the lowest-energy transition is identified as parent transition 1 and the highest-energy transition is identified as 6. The energy difference between parent transitions 1 and 6 is 123 MHz, therefore, at 61.5 MHz the 1n6 and 1p1 sidebands should overlap at the transition center of the spectrum. If the gamma-ray source is stationary, then the energy of the gamma rays emitted differ from the energy of the transition center of the absorber by the isomer shift. Therefore, a stationary source should provide a NFM spectrum of the 1n6 and 1p1 sidebands displaced from 61.5 MHz by plus and minus the isomer shift, respectively [Fig. 5(a)]. The source used was in a Pd lattice, which has an isomer shift of -0.185 mm/s relative to metallic iron. If the source is

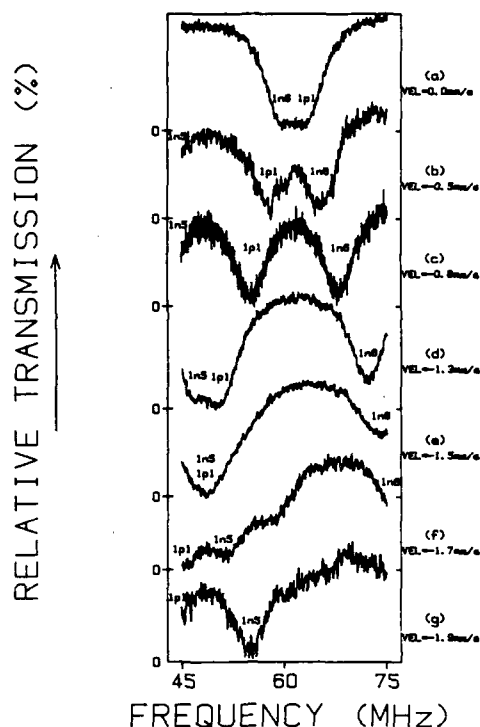


FIG. 5. Typical NFMS data showing first-order sidebands from the highest- and lowest-energy transitions of ^{57}Fe in iron, 1n6 and 1p1, respectively; (a) in the vicinity of the transition center of this Zeeman split absorber. An "n" in the sideband label indicates that the sideband is at a lower energy than its parent transition, whereas a "p" indicates that the sideband is at a higher energy. As the velocity of the source is decreased (b)–(g), the energy of the probing radiation is decreased, and as a result, the sideband from the lower-energy transition, 1p1, appears at lower frequencies. The sidebands from the higher energy transitions, 1n5 and 1n6, appear separated by the excited state splitting frequency of ^{57}Fe in iron, 26 MHz.

then given a constant velocity, the 1n6 and 1p1 sidebands should be displaced from 61.5 MHz by plus and minus (isomer shift – velocity), respectively [Figs. 5(b)–5(g)]. The sign convention is to define a velocity as negative when the source and absorber are moving away from each other. Note that the rf sideband 1n5, which appears in Fig. 5, should be separated from 1n6 by 25.9 MHz, the excited state splitting frequency in metallic iron.

The second set of NFM spectra [Figs. 6(a)–6(c)] were obtained at a lower-frequency range. Higher-order sidebands add together at these lower frequencies and present significant cross sections. These particular spectra are comprised of 24 different sidebands, if one takes into account sidebands out to the fifth order. Following the spectra are computer-generated simulations [Figs. 7(a)–7(c)]. The model, a simple algorithm, shows remarkable agreement with the data. The frequency at which a sideband will appear is

$$F_{j,\text{ord}} (\text{MHz}) = [\text{vel} - (P_j + \text{iso})] (K/\text{ord}), \quad (1)$$

where P_j is the position of the j th parent transition in mm/s, vel is the velocity of the source in mm/s, iso is the isomeric shift between the source and absorber, ord is the order of the sideband, and K is a conversion factor = 11.6 MHz/(mm/s) for the 14.4-keV gamma ray being detected.

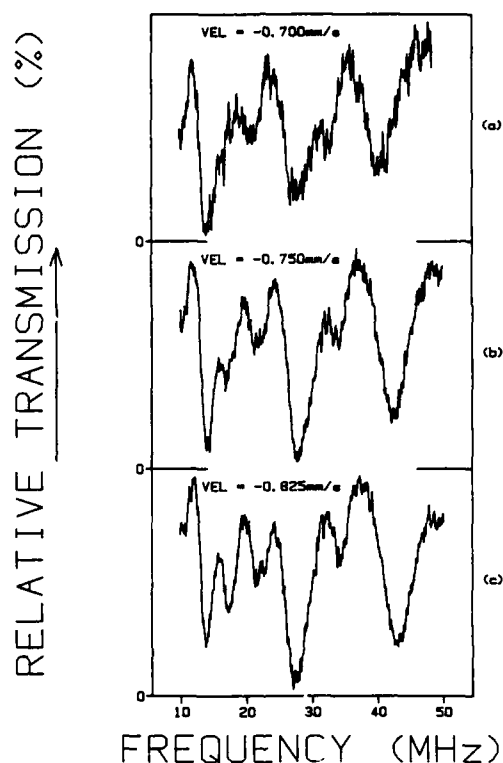


FIG. 6. Nuclear frequency-modulation spectrometer data obtained at lower frequencies has an appearance which belies the underlying complexity of the spectra. Sidebands add together to produce composite sidebands which have amplitudes, widths, and line shapes with a high degree of dependence upon the energy of the probing radiation. As a result, a small change in the source velocity can lead to a significant change in the appearance of a spectrum.

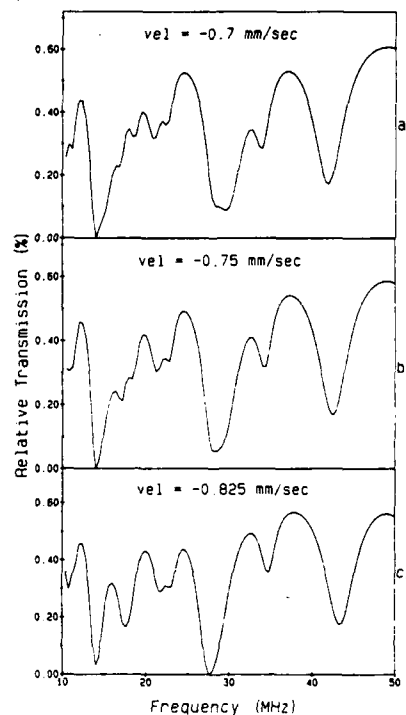


FIG. 7. Computer-generated simulations of the NFM spectra in Fig. 6 obtained from Eqs. (1)–(3). Each simulation is composed of 24 different sidebands.

The linewidth of the sideband as it will appear in the NFM spectrum is

$$\Gamma_{j,\text{ord}} = (\Gamma_{P_j} + \Gamma_g) / \text{ord}, \quad (2)$$

where Γ_{P_j} is the linewidth of the sideband's parent transition and Γ_g is the linewidth of the 14.4-keV gamma ray emitted by the source. The apparent linewidth's dependence on sideband order can be understood by realizing that an n th-order sideband will be displaced by n frequency units, while a first-order sideband is displaced by one frequency unit. Since we were concerned only with the relative amplitudes of a sideband within a given NFM spectrum, the amplitude of a sideband in a spectrum was assumed to be

$$A_{j,\text{ord}} = A_{P_j} / \text{ord}, \quad (3)$$

where A_{P_j} is the amplitude of the sideband's parent transition. By using this equation we have assumed that our spectra were not exhibiting the saturation effects discussed in Ref. 6. This assumption was a convenient mechanism for introducing a sideband amplitude dependence on the relative amplitudes of the parents. For the iron foil used, the relative amplitudes of the parents were taken as 2:2:1:1:2:2 [Fig. 1(a)]. The NFMS is a tool for directly measuring rf sideband position, width, and amplitude. This model shows that the NFMS is a powerful means of indirectly measuring isomeric shifts and the positions and relative amplitudes of the parent transitions.

The NFMS apparatus is versatile. This apparatus is a conventional Mössbauer spectrometer with or without a rf field, as well as a rf sideband spectrometer. A conventional

Mössbauer spectrometer is not capable of measuring rf sidebands when they overlap a parent transition, and resonances have been predicted for certain such overlappings. The NFMS enables one to observe the behavior of rf sidebands in the vicinity of a parent transition, with high enough resolution to discern any fine structure in the sideband which might result from such resonances, because in NFMS, the parent transition appears only as a base line due to its lack of any frequency dependence. With the NFMS, it is also possible to observe directly the effect of the rf field intensity on sideband position, amplitude, and width. It should also be noted that the MCS described in this paper may also be used in a spectrometer similar to the type described by Bolef and Mishory in Ref. 5.

ACKNOWLEDGMENTS

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Appendix II

"Comment on Mössbauer sidebands from a single parent line," by C. B. Collins, P. W. Reittinger, and T. W. Sinor, Phys. Rev. B (submitted).

Comment on "Mössbauer sidebands from a single parent line"

by

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Foils composed of alternating layers of ferromagnetic and nonmagnetic materials immersed in magnetic fields oscillating at radiofrequencies display sidebands on Mössbauer transitions from the nuclei contained in the nonmagnetic regions. Attributed by Chien and Walker [Phys. Rev. B13, 1876 (1976)] to the transfer into the nonmagnetic layer of acoustic phonons excited by magnetostriction in the ferromagnetic layers, this accepted cause of such effects is challenged by new data resulting from a reexamination and extension of that classic experiment.

The paper of Chien and Walker¹ was of such critical importance that it warrants comment over a decade later. Generally perceived as reporting an unarguable proof of a certain basic proposition, it has now been found to have rested upon a demonstrably false assumption. A reexamination of the original experiment shows it to have been so flawed that any conclusions drawn from it must now be considered unproven.

The point of inception had been the original proposal of Mitin^{2,3} that Mössbauer transitions could be excited as part of a multiphoton process in nuclei immersed in intense radiofrequency (rf) fields. In those cases the Mössbauer spectrum was expected to show additional sum and difference frequency lines displaced from the normal lines by integral multiples of the perturbing frequency. In appearance such multiphoton spectra are expected to resemble the transmission spectra which Ruby and Bolef⁴ obtained by imposing periodic Doppler shifts of purely mechanical origin upon the Mössbauer source. This unfortunate similarity in appearance between phenomena arising from such different origins provided the basis for years of critical controversy seemingly resolved by the work of Chien and Walker.¹ The purpose of this comment is to report new data from a repetition and extension of the Chien and Walker experiment that shows their conclusions to be unjustified. Without the force of conviction conveyed by their work, the controversy must be reopened to further investigation.

The earliest experiment in radiofrequency sideband production, reported by Perlow⁵ in 1968, focused upon the components of the 14.4 keV transition in ⁵⁷Fe. Several ⁵⁷Co sources diffused into ferromagnetic hosts were immersed into intense magnetic fields oscillating at radiofrequencies. Those results were explained⁵ as the magnetodynamic modulation of the hyperfine fields and generally conformed to the Mitin hypothesis for multiphoton transitions. Two

of the three groups who initially documented this phenomena favored the magnetodynamic explanation which required no mechanical action^{5,6,7} while the other group began to develop an alternative based entirely upon magnetostriction.^{8,9} Most of the actual experiments had used ferromagnetic hosts to enhance the applied magnetic fields, and such materials are almost invariably magnetostrictive. In the model finally synthesized, periodic Doppler shifts were assumed to be driven by acoustic phonons which were excited by magnetostriction along the greatest dimensions of the material and scattered onto the axis connecting source and absorber. To be effective, this mechanism required the sample to have a large acoustic Q so that displacements of the active nuclei could build to significant values.

Despite the accretion over the years of a large body of phenomenology presumed to describe rf sidebands on Mössbauer transitions, the magnetostrictive-acoustic theory never quantitatively predicted the amplitudes of the sidebands as functions of either applied power or frequency. However, the magnetodynamic models of that time fared no better, and attention turned to "proving" a magnetostrictive origin by distressing the alternative explanations.¹⁰ The obvious difficulty with proving a theory by distressing the alternatives is that those other explanations may not have reached comparable levels of maturation. The magnetodynamic models of the late 60's were relatively easy to destroy.¹⁰ However, the recent successes of ferromagnetodynamics^{11,12} show the early models⁵ of sideband formation to have been inspired, but inadequate approximations. These models simply did not embody the level of sophistication necessary to describe the complex switching behavior of magnetization in ferromagnetic foils subjected to various combinations of static and oscillating fields in those geometries employed.

More recent experiments^{13,14} have shown that the applications of such oscillating magnetic fields to Mössbauer nuclei embedded in nonmagnetic hosts

do produce radiofrequency sidebands by directly modulating the phases of the nuclear states involved in the transitions. However, amplitudes were rather small in those experiments because the driving forces depended only upon the value of applied field, $\mu_0 H$. In 1984, we extended such approaches further by deriving the phase modulation of a nuclear state in a magnetic material.¹⁵ In this case driving forces were proportional to the magnetization $\mu_0 M$ and effects were found to be large.^{15,16,17} It appears that many prior results attributed exclusively to acoustic effects driven by magnetostriction could have also benefited from an unrecognized contribution from direct phase modulations of the nuclear states involved.

From a current perspective it is the experiment reported by Chien and Walker¹ that forms the bulwark of the magnetostrictive-acoustic explanation of Mössbauer sidebands. In that experiment an absorbing foil composed of ferromagnetic and nonmagnetic layers was used to study transport of the causative agent from the ferromagnetic layer into the nonmagnetic region where the sidebands were produced upon Mössbauer transitions of embedded ^{57}Fe nuclei. Very clear evidence showed that the cause did arise in the ferromagnetic Ni layers, producing sidebands in the nonmagnetic stainless steel layers. The most ready explanation at that time was a transport of phonons from one layer to the next with a high acoustic Q. Those experiments were repeated in the work reported here, but with extensions which contradict the classic interpretation of Chien and Walker.¹

Although not unique for all sidebands in a spectrum,¹ the idea of a modulation index m as a measure of the strength of the development of the sidebands offers practical convenience for descriptions. For a magnetostrictive origin,¹

$$m = x_0/\lambda \quad , \quad (1)$$

where x_0 is the amplitude of the periodic displacement of the nuclei and ≈ 0.137 A for the 14.4 keV line of ^{57}Fe . In the corresponding magnetodynamic model,¹⁵

$$m = bH \quad , \quad (2)$$

where H is the applied magnetic field and b provides proportionality between M_s , the saturation magnization of the medium, and H . For relatively small m , the ratio of the magnitude of the first order sidebands to the intensity in the original parent line is proportional to m^2 , which in turn is proportional to P , the applied radiofrequency power.

One of the most compelling results presented by Chien and Walker¹ was a demonstration supposed to show the enhancement of m^2 afforded by tighter acoustic coupling of the layers. They found that electroplating Ni upon a stainless steel foil produced much higher values of m^2 in absorption experiments than could be obtained by gluing a Ni foil to the stainless foil. They attributed the difference to the obviously poorer acoustic properties of the glue. However, as part of this report we observe that their stainless steel foil was electroplated on both sides with Ni while the epoxied bond was used to join a single Ni foil to one side of the stainless absorber. While the m defined by Eq. (1) for a single foil could not be additive if produced in different magnetostrictive layers, in principle the M_s upon which m depends in Eq. (2) could add coherently. Two sources of m arising from distinctly separate sources could give a resulting modulation of $4m^2$ in a magnetodynamic model. Chien and Walker failed to recognize¹ that even in the magnostriuctive model two sources of m generated in the two electroplated layers should give a modulation index of $2m^2$ in the absorber foil. Instead, they attributed the increased sideband intensity developed by the two plated sources in comparison to the one glued source only to the advantage they assumed for a plated contact over a glued interface. They reported no comparison of the effects of

gluing or plating the *same number of ferromagnetic layers to the absorber foil*. Reported here is a repetition of the Chien and Walker experiment which showed that the effect of two foils varied from two to four times that produced by a single foil joined in the same fashion, depending upon the static magnetic bias applied.

In our experiment the absorber was a $2.5\text{ }\mu\text{m}$ paramagnetic stainless steel (SS) foil with 90.6% enrichment of ^{57}Fe . For the nonabsorbing ferromagnetic drivers, $2.5\text{ }\mu\text{m}$ Ni foils were used, all of which were cut from a single sheet of polycrystalline Ni. The stainless-steel absorber was sandwiched between two Ni foils and held in rigid contact by mounting the foils between glass cover slides of $100\text{ }\mu\text{m}$ thickness. A conventional Mössbauer spectrometer, modified for rf experiments, Fig. 1, utilized a 25 mCi source in a Rh matrix to obtain the ^{57}Fe absorption spectra. The 14.4 keV gamma rays were detected with a Kr gas filled proportional counter biased with 1.8 kV.

A 25 MHz rf magnetic field was applied by mounting the foils in the cylindrical induction coil of an L-C tank circuit. In obtaining data for a direct comparison between the effect of one Ni driver versus two, the product of the applied rf power P and the electrical Q of the circuit containing the rf induction coil was maintained at constant values. Elementary analysis shows that if PQ is constant the rf current in the coil of such a circuit is also constant and hence the two absorber arrangements are subjected to applied fields of the same intensity H . The results of the first experiment verified the linearity of the first order sideband amplitudes at 25 MHz for SS with two Ni drivers with PQ products of 75, 150, and 300 W Fig. 2. The spectra are scaled so that the intensity of the central Mössbauer absorption peak of ^{57}Fe in SS is held constant in order to make direct comparisons of the sideband amplitudes.

Having established the linearity of the first order sidebands in the Ni-SS-Ni sandwich, one of the Ni drivers was removed and the experiment was repeated with the same PQ products as before. Figure 3 shows a comparison of the sideband amplitude for two Ni drivers versus one; in this configuration two Ni drivers give twice the effect of one driver foil.

In the next experiment a comparison between the effect of one source of excitation with that from two sources when both were biased with a static magnetic field. Rare earth magnets were placed about the induction coil such that the static magnetic field was mutually orthogonal to the rf magnetic field and the direction of gamma-ray propagation.

The linearity of the sideband amplitudes at 25 MHz as a function of PQ was again established (Fig. 4) to insure that the introduction of the static magnetic field did not introduce any nonlinearities to the system. The scale thus established was used to measure the decrease in the sideband amplitude when one of the sources of excitation was removed from this biased sandwich. As is clearly shown in Fig. 5, the sideband amplitudes obtained with two driver foils are four times the amplitudes obtained with one driver foil. Therefore, with the application of a static B-field, two sources of excitation give four times the effect.

The results of this reexamination of the Chien and Walker experiment support only the first conclusion reached in that original work, namely that the causative agent of rf sidebands can be produced in a ferromagnetic layer and then transported into a nonmagnetic layer. Their other conclusion is completely refuted by this demonstration because the effects they attributed to the type of coupling between layers most probably resulted from the relative numbers of magnetic and nonmagnetic layers.

These new results go beyond the propositions tested by Chien and Walker¹ and display behaviors completely inconsistent with the traditional magne-

to restrictive-acoustic origin of Mössbauer sidebands. In experiments such as these, acoustic phonons are the bosons associated with vector fields driven by tensor forces, *not vector forces*. Without invoking stimulated emission, we can conceive of no way in which tensor sources which are physically separated can produce coherent vector fields in a space between them, even if they are temporally synchronized. The stimulated emission of phonons to produce coherent additions of the displacements arising from the different sources would imply the existence of a threshold of power, above which two modulation indices of m would give an effect of $4m^2$ and below which only $2m^2$. No such threshold was suggested by data similar to that of Fig. 5 which was obtained over an adequate range of powers.

In view of the growing number of successes of the model for the direct modulation of the phases of the nuclear states and these new results which question the validity of the conclusions of the Chien and Walker¹ experiment, it would appear that the controversy over the origin of Mössbauer sidebands must be reopened.

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CAPTIONS

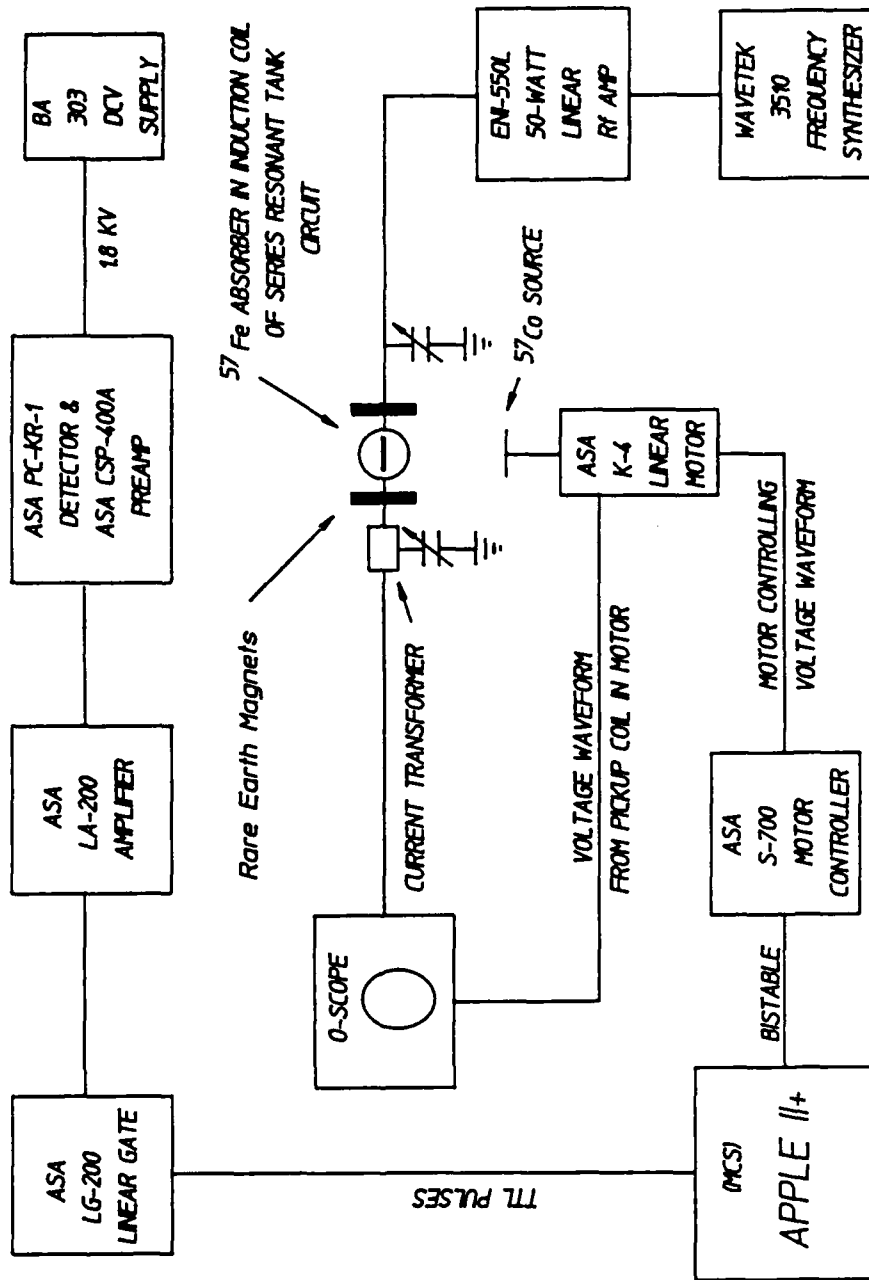
Fig. 1: Schematic drawing of the experimental arrangement used.

Fig. 2: Experimental verification of the linearity of the first order sidebands at 25 MHz as a function of the applied rf power. The product of the applied rf power, P , and the quality factor, Q , of the circuit are used to insure reproducibility of the rf field strengths.

Fig. 3: Comparison of first order sideband amplitudes for one Ni driver foil versus two at 25 MHz with a PQ product of 300 W.

Fig. 4: Establishment of linearity of the first order sidebands at 25 MHz with $PQ = 75, 150$ and 300 W when the foils are biased with a static B-field.

Fig. 5: Comparison of sideband amplitudes for one driver foil versus two when both are biased by a static B-field with $PQ = 300$ W. Here two foils give four times the effect of one thus giving a modulation index of $4m^2$.



Linearity of First Order Sidebands as a Function of Rf Power

PQ = 75 W

PQ = 150 W

PQ = 300 W



Ni



SS

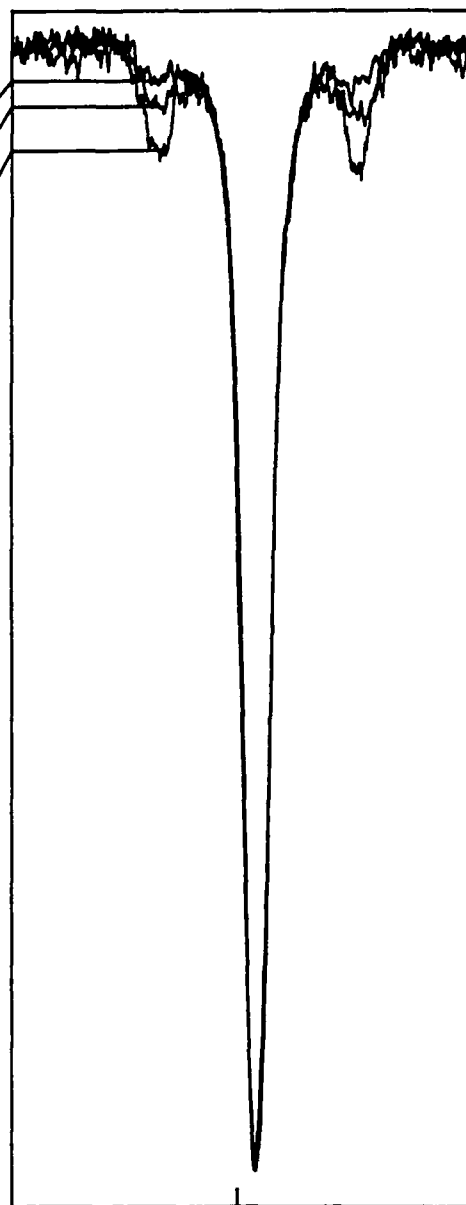


Ni

Rf

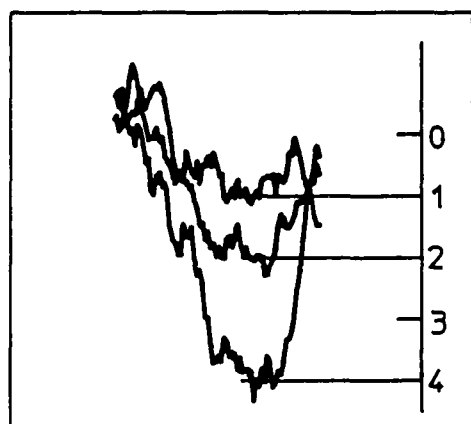
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% Transmission



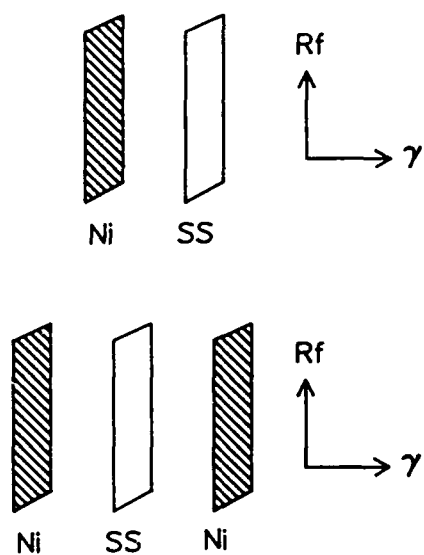
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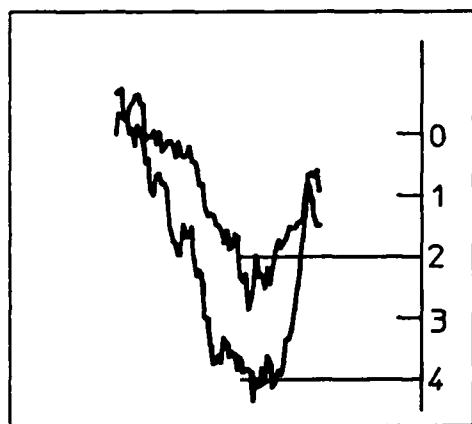
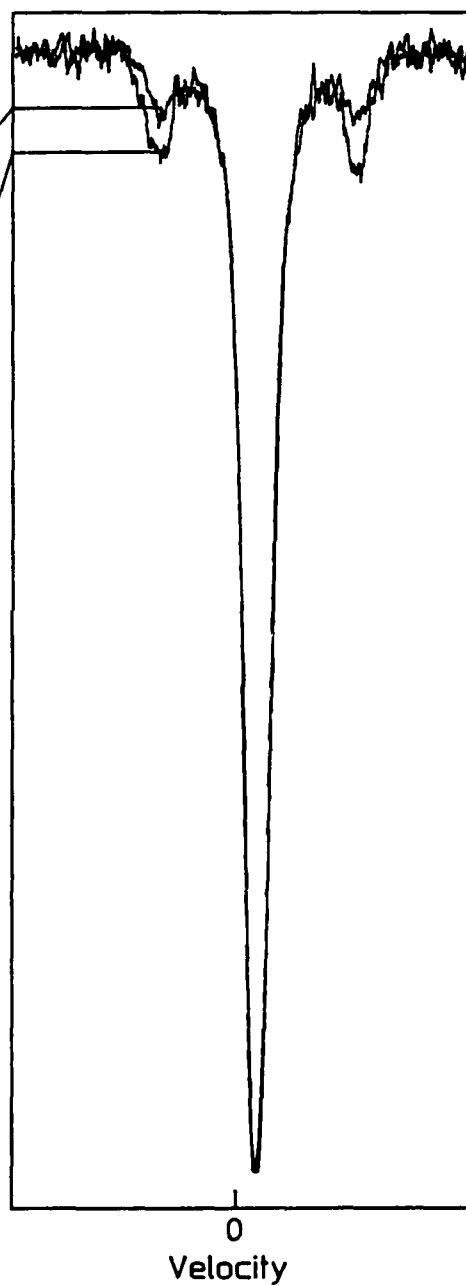


Details of First Order Sidebands

Comparison of Sideband Amplitudes for One Driver Foil vs Two PQ = 300 W



% Transmission



Details of First Order Sidebands

Linearity of First Order Sidebands Under the Influence of a Static B-Field

PQ = 75 W

PQ = 150 W

PQ = 300 W



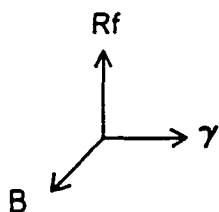
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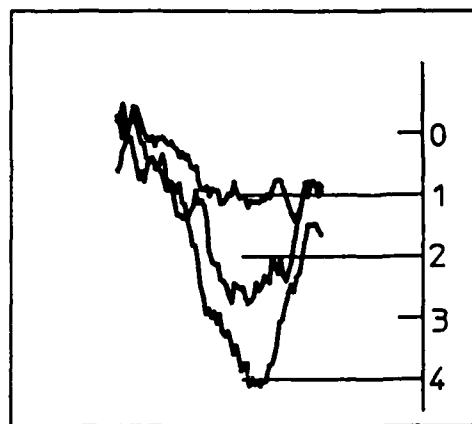
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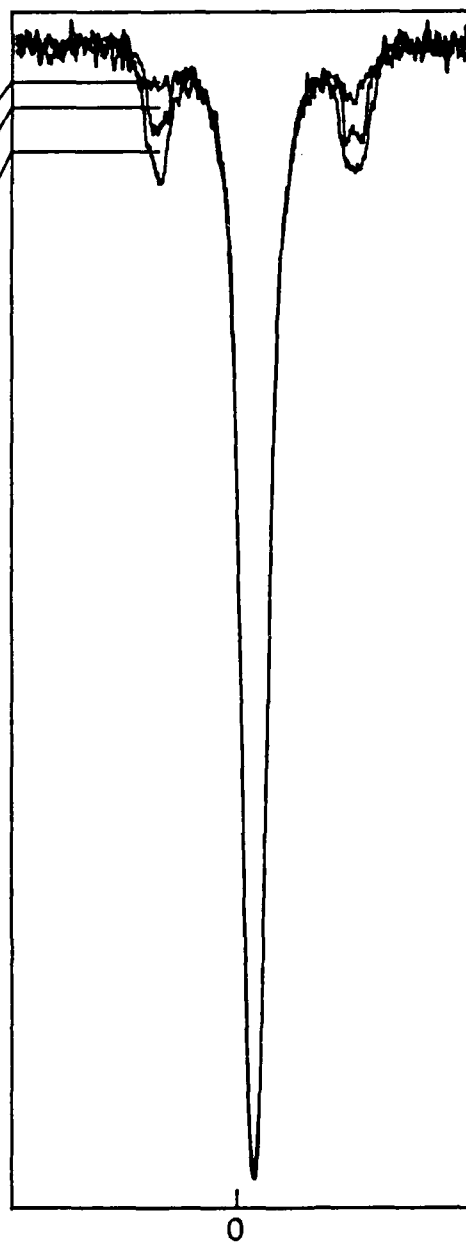
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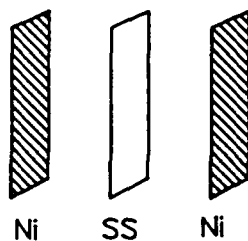
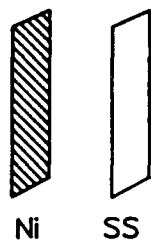


Details of First Order Sidebands

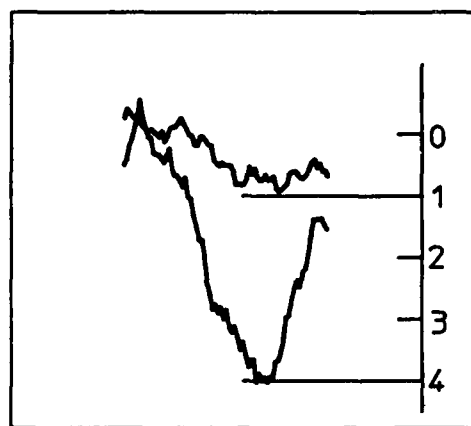


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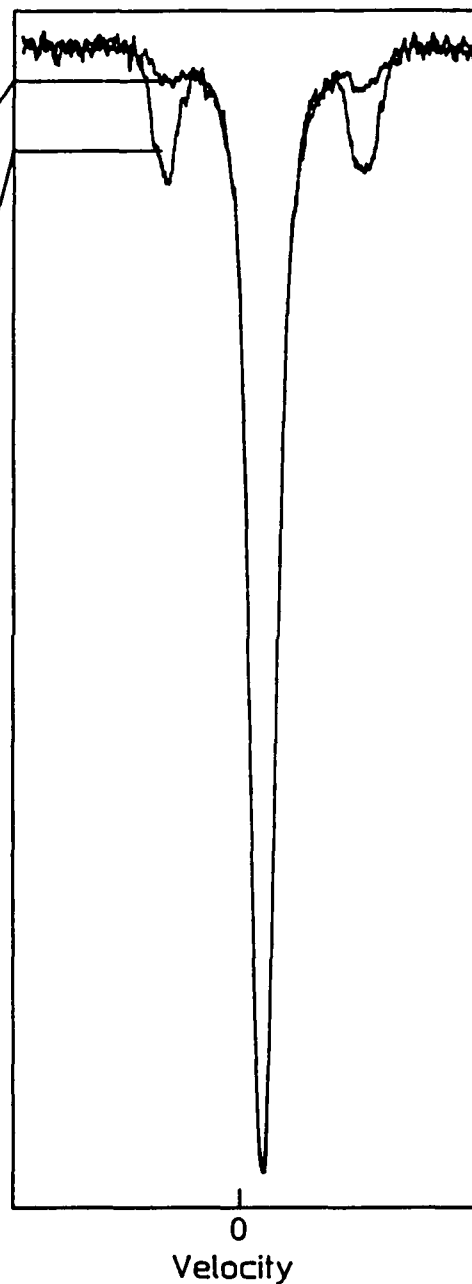
Comparison of Sideband Amplitudes for
One Ni Driver vs Two with a Static
B-Field Applied; PQ = 300 W



% Transmission



Details of First Order Sidebands



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